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(19) THE PATENT OFFICE LONDON

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(54) FLUID-WALL REACTORS AND THEIR UTILIZATION IN HIGH TEMPERATURE CHEMICAL REACTION PROCESSES

(71) We, THAGARD TECHNOLOGY COMPANY, a limited partnership organised and existing under the laws of the State of California, of 2712 Kelvin Avenue, Irvine, State of California, United States of America, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The present invention relates to fluid-wall reactors for high temperature chemical reaction processes, as well as to the various processes which may be conducted in such reactors, many of which processes previously have been impractical or only theoretically possible. Both the fluid-wall reactor and the processes employed in such reactors utilize radiation coupling as a heat source and maintain the contemplated chemical reactions in isolation within a protective fluid blanket or envelope out of contact with the containing surfaces of the reactor.

The subject-matter of the present application is closely related to that of our co-pending application No. 40886/75 (Serial No. 1,530,314).

High temperature reactors are presently employed to carry out pyrolysis, thermolysis, dissociation, decomposition and combustion reactions of both organic and inorganic compounds. Substantially all such reactors transfer heat to the reactants by convection and/or conduction, but this characteristic inherently produces two major problems which limit the nature and scope of the reactions which may be carried out. Both problems result from the fact that in a conventional reactor which transfers heat to the reactants by convection, the highest temperature in the system is necessarily at the interface between the inside wall of the reactor and the reactant stream.

The first problem involves the limitations on available temperatures of reaction which are imposed by the strength at elevated temperatures of known reactor wall materials. The decreasing capability of such materials to maintain their integrity under conditions of increasing temperature is, of course, well known. However, since it is necessary that such materials be heated in order that thermal energy may be transferred to the reactant stream, available reaction temperatures have been limited by the temperature to which the reactor wall could be safely heated. This factor is particularly critical in cases where the contemplated reaction either must take place at or produces high pressures.

The second problem inherently results both because the wall of a conventional reactor is at the highest temperature in the system and because convective/conductive heat transfer requires contact between the wall and the reactant stream. Being at such elevated temperature, the reactor wall is an ideal if not the most desirable reaction site in the system and, in many instances, reaction products will accumulate and build up on the wall. Such buildup impairs the ability of the system to transfer heat to the reactants and this ever increasing thermal impedance requires the source temperature to be raised progressively just to maintain the initial rate of heat transfer into the reactant stream. Obviously, as the build-up increases, the required source temperature will eventually exceed the capabilities of the reactor wall material. Moreover, as additional energy is required to sustain the reaction, the process becomes less efficient in both the technical and economic sense. Thus, at the point where the contemplated reaction can no longer be sustained on the basis of either heat transfer, strength of materials, or economic considerations, the system must be shut down and cleaned.

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Usually, cleaning is performed mechanically by scraping the reactor wall or chemically by burning off the deposits. In some continuous processes, it has been attempted to scrape the reactor wall while the reaction proceeds. However, the scraping tool itself necessarily gets hot, becomes a reaction site and, thereafter, must be cleaned. In any event, this down time represents a substantial economic loss. In many instances, a second system will be installed in order to minimize lost production time. However, such additional equipment generally represents a substantial capital investment. Some high temperature chemical reactors include a tube which is heated to a temperature at which its inner walls emit sufficient radiant energy to initiate and sustain the reaction. However, as in the case of conductive and convective reactors, for reactions yielding solid products there is frequently an undesirable build-up of product on the tube walls which leads to reduced heat transfer and even clogging of the tube.

The reactor disclosed in U.S. patent No. 2,926,073 is designed to produce carbon black and hydrogen by the pyrolysis of natural gas. The process is stated to be continuous but, in practice, the convective heat transfer principle under which the reactor operates causes serious problems both in sustaining and controlling the reaction. Since the heated tubes of the reactor are ideal reaction sites, carbon invariably builds up and eventually clogs the system. More serious, however, is the problem of thermal runaway which can result in explosions. With respect to this condition, it has been determined that during pyrolysis of natural gas, thermal conductivity of the gas phase suddenly increases from about five to thirty times, depending upon the composition of the gas. Because the temperatures in a conventional convective reactor cannot be regulated with sufficient speed and accuracy to compensate for this phenomenon, in some instances the system would become unstable and explosions would result. Such conditions are inherent in conventional reactors and, as yet, no way has been found to overcome this problem.

U.S. patent No. 3,565,766 represents a recent attempt to upgrade coal by pyrolysis. The disclosed system comprises a series of hollow steel vessels which act as multi-stage fluidized beds at successively increasing temperatures up to about 1600°F. Fluidization at lower temperatures is achieved by an inert gas which may itself supply heat although external heating is contemplated. At higher temperatures, fluidization is achieved by the overhead gas obtained in the final stage; and, in the final stage, temperature is maintained by internal combustion of the char in air or oxygen. Because it relies primarily upon heat transfer by convection, this system is subject to many of the defects and disadvantages which have previously been discussed.

The apparatus for the manufacture of carbon black disclosed in U.S. patent No. 2,062,358 includes a porous tube disposed within a heating chamber. Hot gas is directed from a remote furnace into the chamber, and thereafter forced through the wall of the porous tube to mix with the reactants. Thus, only convective transfer of heat from a fluid to reactants is employed. This, necessitates the flow of a large volume of fluid through the heating chamber in order to make up for heat losses.

U.S. patent No. 2,769,772 discloses a reactor for heat-treating fluid materials such as hydrocarbons which includes two concentric tubes disposed in a flame heated furnace. Reactants flow axially through the pervious inner concentric tube. A heat-carrier gas flowing in the annular chamber between the concentric tubes is heated by contact with the outer wall. Fluids in the inner tube are heated by convection when the heat-carrier gas passes through the pervious wall and mixes with them. Radiant heat transfer is expressly avoided. In fact, it is impossible to heat the inner tube without simultaneously heating the outer tube to at least as high a temperature.

The surface-combustion cracking furnace of U.S. patent No. 2,436,282 employs the convective heat carrier gas principle similar to that of U.S. patent No. 2,769,772. The furnace includes a porous, refractory tube enclosed by a jacket. A combustible fluid from an annular chamber is forced through the porous wall to the inside of the tube where it is ignited. It is evident, however, that the combustible fluid in the annular chamber will explode unless it is forced through the porous wall at a rate faster than the rate of flame propagation back through the wall. Likewise, the temperature in the annular chamber must be maintained below the ignition temperature of the gas/air mixture. Combustion products from the surface flame mix with reactants in the furnace diluting and possibly reacting with them. Heat is imparted to the reactants by convective mixing of the combustion products and the reactants.

U.S. patents Nos. 2,670,272; 2,670,275; 2,750,260; 2,915,367; 2,957,753, and 3,499,730 disclose combustion chambers for producing pigmentary titanium dioxide by burning titanium tetrachloride in oxygen. In the '275 patent which is representative of this group, titanium

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tetrachloride is burned in a porous, refractory tube. An inert gas is continuously diffused through the porous tube into a combustion chamber where it forms a protective blanket on the inner surface of the tube. This gaseous blanket substantially reduces the tendency of the titanium dioxide particles to adhere to the walls of the reactor. Since the combustion of titanium tetrachloride is an exothermic reaction, no provision is made to supply heat to the reaction mixture as it passes through the tube. In fact, the '275 patent teaches that it is advantageous to remove heat from the reactor chamber either by exposing the porous tube assembly to the atmosphere or by circulating a cooling fluid through a coil disposed about the porous tube.

The present invention provides a process for carrying out a chemical reaction at an elevated temperature, wherein one or more reactants are situated in a reaction zone defined by a fluid envelope of generally annular cross-section within a reactor tube, and radiant energy generated outside the reactor tube is collected and focussed onto the reactant or reactants in the reaction zone, the wall of the reactor tube and the fluid envelope both being substantially transparent to the radiation used, sufficient radiant energy being absorbed in the reaction zone to raise the temperature of the or at least one reactant to sustain the chemical reaction.

The invention also provides a reactor for carrying out a chemical reaction at an elevated temperature, comprising a reactor tube having an inlet end and an outlet end, means for generating an envelope of fluid to define a fluid-walled reaction zone within the reactor tube, a source, located outside the reactor tube, of radiant energy to which the wall of the reactor tube is substantially transparent, and means for collecting and focussing onto the reaction zone, in operation sufficient radiant energy from the source to raise the temperature of one or more reactants situated there to sustain a chemical reaction.

The process of the invention is advantageously carried out as follows. An annular envelope of an inert fluid which is substantially transparent to radiation is generated, the envelope having a substantial axial length, Next, at least one reactant is passed through the core of the envelope along a predetermined path which is substantially coincident with the envelope axis, the reactants being confined within the envelope. After the reactant flow has started, high intensity radiant energy is directed through the envelope to coincide with at least a portion of the path of the reactants. Sufficient radiant energy is absorbed in the core to raise the temperature of the reactants to a level required to sustain the desired chemical reaction.

In the event that the reactants are themselves transparent to radiant energy, an absorptive target may be introduced into the reactant stream, or introduced prior to the introduction of the reactants. The target will absorb sufficient radiant energy to raise the temperature in the core to the desired level. In some instances, however, while the reactants are transparent to radiation, one or more of the reaction products will be an absorber. In such event, once the reaction has been initiated the target may be withdrawn and the reaction will continue. An example of such reaction is the pyrolysis of methane to carbon and hydrogen.

Some reactions will reverse either partially or completely if the reaction products are not cooled immediately and rapidly. In such cases, it is further contemplated that cooling of reaction products and any remaining targets to prevent such undesired chemical reactions be carried out immediately upon completion of the desired reaction.

The high temperature fluid-wall reactor of the present invention transfers substantially all of the required heat to the reactants by radiation coupling, either directly or via a target. The reactor comprises a tube having an inlet and an outlet end, the interior of the tube defining a reaction zone. The reactor preferably includes means for introducing an inert fluid into the reactor chamber to provide a protective blanket for the radially inward surface of the reactor tube, and means for introducing at least one reactant into the reactor tube through the inlet end to cause such reactants to be directed in a predetermined path axially of the reactor tube. The inert fluid blanket confines the reactants substantially centrally within the reactor tube and out of contact with the reactor tube walls. High intensity radiant energy is generated and directed into the reactor chamber to coincide with at least a portion of the path of the reactants, sufficient radiant energy being absorbed to raise the temperature of the reactants to a level required to sustain the desired chemical reaction.

In contrast to the conventional convective reactors, the present invention relies upon radiation coupling to transfer heat to the reactant stream. The amount of heat transferred is independent both of physical contact between the reactor wall and the stream and of the degree of turbulent mixing in the stream. The primary consideration for heat transfer in the

present system is the radiation absorption coefficient (α) of the reactants. The inert fluid blanket which protects the reactor wall is substantially transparent to radiation and thus exhibits a very low value of (α). This enables radiant energy to be transferred through the blanket to the reactant stream with little or no energy losses. Ideally, either the reactants themselves or a target medium will exhibit high (α) values and will thus absorb large amounts of energy, or alternatively, the reactants may be finely divided (as in a fog) such that the radiation is absorbed by being trapped between the particles. Since materials which are good absorbers are generally good emitters of radiation, when the reactants or targets are raised to a sufficiently high temperature, they become secondary radiators which re-radiate energy throughout the entire reacting volume and further enhance the heat transfer characteristics of the system. This occurs almost instantaneously and is subject to precise and rapid control. Moreover, the re-radiation phenomenon which ensures rapid and uniform heating of the reactants is completely independent of the degree of turbulent mixing which may exist in the reactant stream.

The present high temperature chemical process and apparatus provide a solution to problems which have plagued the art and thus permit the carrying out of reactions which heretofore have been impractical or only theoretically possible. Because heat is supplied by radiation coupling rather than by convection and/or conduction, the temperature of the reactant stream may be independent of both the temperature of the reactor wall and of the condition of the reactant stream, and the serious strength of materials problem is overcome. Two embodiments of the present reactor, described in more detail below, contemplate that the reactor wall in fact be cooled. The temperature within the reaction zone, however is preferably at least 2300°F.

The use of radiation coupling further enables the accurate and almost instantaneous control of heat transfer rates which is impossible to achieve in a conventional convective reactor. Furthermore, the present reactor may provide a power flux at the reaction site in excess of 10,000 watts/cm^2; preferably the power flux in the reaction zone is at least 180 watts/cm^2. Even this lower figure represents a great improvement over the 2—3 watts/cm^2 which is ordinarily obtained in conventional reactors.

The reactions which may be carried out by the process of this invention as implemented by the present reactor are many and varied. For example, organic compounds, particularly hydrocarbons, may be pyrolyzed to produce carbon and hydrogen without the attendant build-up and thermal runaway problems which were encountered in the prior art. Saturated hydrocarbons may be partially pyrolized to obtain unsaturated hydrocarbons; thus, for example, propane and ethane may be dehydrogenated to propylene and ethylene, respectively. Unsaturated hydrocarbons may be partially pyrolyzed in the presence of hydrogen to form saturated hydrocarbons and, more specifically, petroleum products may be thermally cracked. Thus, gas oil may be readily converted into diesel oil, kerosene, gasoline fractions or even methane. Halogen intermediates may be added to partially pyrolyzed hydrocarbons to produce higher molecular weight compounds. Hydrocarbons may be completely or incompletely pyrolyzed in the presence of steam to form carbon monoxide and hydrogen; additional hydrogen may then be added and the reaction carried out to form alkane series hydrocarbons which are high BTU-value fuel gases.

Inorganic compounds may likewise be pyrolyzed. For example, salts or oxides of iron, mercury, silver, tungsten and tantalum, among others, may be dissociated to obtain pure metals. Oxides of iron, nickel, cobalt, copper and silver, to name a few, may be directly reduced in the presence of hydrogen with the same result. This list is by no means intended to be exhaustive.

Novel composite products may also be produced by the present process. For example, carbon or talc particles coated with silicon carbide may be obtained. This product serves as an excellent abrasive because as it is used, it continually breaks up and forms fresh new sharp surfaces. Particles of certain elements such as U^{235} may also be encapsulated in a chemically-tight envelope of another material such as carbon; this particular product is useful as a nuclear reactor fuel element.

It is further contemplated that the present invention may provide the terminal step in conventional aerobic incineration of waste such as garbage and sewage. The relatively low temperatures encountered in current incineration processing techniques permit the formation or organic peroxides and oxides of nitrogen which are major contributors to photochemical smog and other forms of air pollution. Because such compounds are not stable at the higher processing temperatures afforded by the present invention, a waste incineration effluent which is very low in pollutants may be obtained.

Further, the present invention contemplates the high temperature anaerobic destructive distillation and/or disassociation of waste to yield useful products such as carbon black, activated charcoal, hydrogen, and glass cullet, to name a few. The addition of steam to carbonaceous waste will produce carbon monoxide and hydrogen which may then be processed in the conventional manner to obtain fuel gases. Finally, the addition of hydrogen to carbonaceous waste will produce petroleum-equivalent heavy oils and other petroleum products. Thus, substantial reductions in air pollution as well as significant economic gains may be realized through such contemplated applications of the present invention.

The present invention represents a major breakthrough in the art. Because it makes available for the first time a source of thermal energy which has never been harnessed in this manner, its potential applications are numerous and varied. Moreover, in surmounting the strength of materials problem which has shackled the art for many years, this invention makes possible in the practical sense many useful chemical reactions which have long been known but which could not be performed because of temperature limitations inherent in reactors which depended upon convective and/or conductive heat transfer.

The invention will now be described in further detail, by way of example, only with reference to the accompanying drawing, which represents an elevation in partial section of one embodiment of the reactor of the present invention.

Referring to the drawing, a high-temperature chemical reactor 10 comprises a reactor tube 11 which has an inlet end 12 and an outlet end 14. The reactor tube 11 includes an inner wall 15 and an outer wall 16 which define an annular channel therebetween and the interior of the tube 11 constitutes a reactor chamber 17. The tube 11 is made of a material which is substantially transparent to radiation. Suitable materials of this nature which exhibit a very low absorption coefficient (α) include glass, quartz, hot sintered aluminium oxide, hot sintered yttrium oxide, borosilicate glass, for example, pyrex (Trade Mark), silicate glass, for example, Vycor (Trade Mark), and sapphire; organic polymers such as Plexiglass (Trade Mark), (acrylic), Lucite (Trade Mark) (acrylic), polyethylene, polypropylene and polystyrene; and, inorganic salts such as the halides of sodium, potassium cesium lithium or lead.

As used herein, the terms "radiant energy" and "radiation" are intended to encompass all forms of radiation including high-energy or impacting nuclear particles. However, because the practical use of such radiation is not possible under the present state of the art, electro-magnetic radiation, particularly of wavelengths ranging from 100 microns to 0.01 microns, is considered to be the primary energy source upon which design considerations are to be based.

During operation of the reactor 10 a fluid medium which is substantially transparent to radiation is introduced through inlet 18, circulates throughout the annular channel to cool the reactor tube 11 and exits through outlet 19. Such fluid medium may be a gas or a liquid; representative suitable fluids which have low coefficients of absorption (α) include liquid or gaseous water, heavy water, nitrogen, oxygen and air.

Means for introducing an inert fluid into the reactor chamber 17 through an inlet 20 comprises first and second laminar diffusers 21 and 22, respectively, which are disposed adjacent the inlet end 12 of the tube 11. Such diffusers 21, 22 may be in the form of honeycomb cores or any other suitable configuration which causes a fluid directed under pressure therethrough to flow in a substantially laminar fashion. The inert fluid is thus introduced substantially axially into the reactor chamber 17 to provide a protective blanket for the radially inward surface of the reactor tube 11 and is collected for recirculation as it exits through outlet 23. The inert fluid is substantially transparent to radiation in that it has a low (α) value. Fluids which are suitable for this purpose include simple gases such as helium, neon, argon, krypton and xenon; complex gases which do not decompose to form a solid product such as hydrogen, nitrogen, oxygen and ammonia; and, liquid or gaseous water. The term "inert" as used herein, involves two factors: the ability of the fluid to react chemically with the material of the reactor tube 11 and the ability of the fluid to react chemically with the materials which are being processed. Thus, the selection of an "inert" blanket fluid depends in each instance upon the particular environment. Except as otherwise specifically provided, it is desirable that the fluid be inert with respect to the reactor tube and it is usually desirable that the fluid be inert with respect to the reaction which is carried out. However, it is contemplated that in some instances the "inert" fluid of the protective blanket shall also participate in the reaction as, for example, where iron or carbon particles are reacted in the presence of a stream blanket to produce iron oxide and hydrogen or carbon monoxide and hydrogen, respectively.

Reactants are introduced into the reactor chamber 17 through an inlet 24 at the inlet end 12 of the reactor tube 11. The reactants are directed along a predetermined path 25 axially of the reactor tube 11 and are confined by the protective inert fluid blanket substantially centrally within the reactor chamber 17 out of contact with the reactor tube 11.

A high-intensity radiant energy source (not shown) is disposed within a polished reflector 31 which is mounted on a frame 32 externally of the reactor tube 11. The radiant energy source may be a plasma arc, a heated filament, a seeded flame, a pulsed flashlamp or other suitable means; a laser may also serve as the source but, at present, laser technology has not been sufficiently developed to the extent where it is economically practical for the purposes contemplated by the present invention. The radiant energy generated by the source is collected by the reflector 31 and is directed through the tube 11 into the reactor chamber 17 to coincide with at least a portion of the path 25 of the reactants. Sufficient radiant energy will thus be absorbed to raise the temperature of the reactants to a level required to initiate and carry out the desired chemical reaction. As previously stated, the tube 11, the cooling fluid and the inert blanket are all substantially transparent to radiant energy. Accordingly, they do not interfere to any great extent with the transmission of energy to the reactant stream (stream) and remain relatively cool. Thus, the reactor tube 11 is not subjected to appreciable thermal stress and remains free from precipitates and other deposits which would normally accumulate.

The above discussion presumes that the reactants themselves exhibit a relatively high radiation absorption coefficient (α). However, if such is not the case, a radiant energy absorptive target must be introduced into the reactor chamber 17 coincident with at least one point along the path 25 of the reactants on which the radiation is incident. In the embodiment shown in the drawing, the target medium is a finely divided solid such as carbon powder or other suitable material which enters the reactor chamber 17 together with the reactants through inlet 24 and absorbs sufficient radiant energy to raise the temperature of the reactants to the required level.

Alternatively, the target may be a liquid such as tar, asphalt, linseed oil or diesel oil, and may include solutions, dispersions, gels and suspensions of varied make-up which may be readily selected from available materials to suit particular requirements. The target may be a gas which preferably exhibits absorption in the electromagnetic spectrum from 100 microns to 0.01 microns; such gases include ethylene, propylene, oxides of nitrogen, bromine, chlorine, iodine, and ethyl bromide. The target may also be a solid element made of a material such as carbon which is disposed in the reactor chamber 17 along at least a part of the path 25 of the reactants where the radiation is incident.

Other means for raising the temperature of the reaction to the required level may include an electrically heated element, an electric arc or a flame disposed within the reactor chamber 17 coincident with at least a part of the path 25 of the reactants at or before the incidence of the radiation. In such instances, the initiating heat source is self-contained and is not derived from the radiant energy generating means. Such means are particularly useful where the reactants themselves are transparent to radiation but at least one of the reaction products is an absorber. Thus, once the contemplated reaction has been initiated, the temperature raising means may be deactivated because the reaction products will absorb sufficient radiant energy to sustain the reaction. Likewise, if a target medium is used, it may be discontinued or withdrawn once the reaction has begun as by operation of a control means 35. An example of a reaction where a target or other initiating means is required only at the outset is the pyrolysis of methane to produce carbon and hydrogen.

As previously stated, some reactions will reverse either partially or completely if the reaction products are not cooled immediately and rapidly. For this purpose, reaction product cooling means 40 may be provided with the reactor chamber 17 adjacent the outlet end 14 of the reactor tube 11. One embodiment of such means 40 is disposed substantially centrally within the reactor chamber 17 and includes a tubular member 41 having an internal channel through which is circulated a coolant such as water. The radially inward surface of the tube 41 is designed to constitute an absorber of radiant energy. As the reaction products, remaining reactants and targets, if any, pass within the cooled tube 41, heat is transferred rapidly by radiation coupling and the system is effectively quenched to prevent any further undesired chemical reactions.

The present invention further contemplates a second embodiment in which the reactor tube may be constructed of a porous material which is substantially transparent to radiation. Suitable wall materials include for example porous quartz, porous glass frit, and porous sapphire. An inert fluid which is

substantially transparent to radiation may thus be introduced into the reactor chamber radially inwardly through the porous reactor wall rather than axially in a laminar fashion as described with respect to the first embodiment. Radiant energy is generated, collected and directed into the reactor chamber also as described with respect to the first embodiment.

As previously mentioned, high temperature chemical reaction processes conducted in accordance with the present invention necessitate the use of a fluid wall substantially transparent to the radiation employed, the wall preferably being in the form of an annular envelope or blanket having a substantial axial length. The annular envelope may be generated in a direction generally parallel to its axis or in a direction generally perpendicular to its axis and radially inwardly of its outer circumferential surface.

In the former instance, as previously described with respect to the first embodiment of the present reactor, the envelope fluid must be maintained in laminar flow to prevent intermixing with the reactant stream. This requirement imposes certain limitations upon the axial length of the envelope because such laminar flow, and thus blanket integrity cannot be maintained for indefinite lengths downstream, especially if a particularly violent reaction is contemplated. Accordingly, this manner of generating the envelope is most suitable for smaller-scale and laboratory applications.

In the latter instance, as previously described with respect to the second embodiment of the present invention, the integrity of the fluid envelope is independent of any flow considerations and may be maintained for an axial distance much greater than that obtainable in the case of the axially injected laminar envelope. The primary requirement is to maintain the flow of the inert fluid under a greater pressure than that of the reactant stream to prevent the reactants from "punching through" or otherwise breaking out of confinement within the envelope.

After the envelope has been generated, the process of the invention is preferably conducted as follows. At least one reactant is passed through the core of the envelope along a predetermined path which is substantially coincident with the envelope axis. The envelope confines the reactants therewithin and out of contact with the containing surfaces of the reactor chamber.

Finally, high intensity radiant energy is directed into the envelope core to coincide with at least a portion of the predetermined path of the reactants, and sufficient radiant energy is absorbed in the core to raise the temperature of the reactants to a level required to sustain the desired chemical reaction.

In the event that the reactants will not themselves absorb radiant energy, an absorptive target may be introduced along the path of the reactants. The target will then absorb sufficient radiant energy to raise the temperature in the core to the level required to initiate the desired chemical reaction. As previously stated, if the contemplated reaction is such that the transparent reactants produce at least one product which absorbs radiant energy, the target may be withdrawn, or its supply discontinued, after the reaction has been initiated.

The contemplated process may further include the step of cooling the reaction products and any remaining reactants and/or targets immediately after the desired reaction has been contemplated. The purpose of this procedure is to terminate the desired reaction and to prevent the occurrence of any further undesired reaction. The products, targets and remaining reactants may be cooled conveniently and effectively by radiation heat transfer to a cool, radiant energy absorbing surface.

The fluid-wall reactors of the invention may be used in virtually any high temperature chemical reaction, many of which reactions have been previously regarded as either impractical or only theoretically possible. The most important criterior for utilizing these fluid-wall reactors in a particular high temperature chemical reaction is whether such reaction is thermodynamically possible under the reaction conditions. Utilizing these fluid-wall reactors, such high temperature chemical reaction processes can be conducted at temperatures up to about 6000°F.

Among the reactions which may be carried out in the fluid-wall reactors of the invention are the dissociation of hydrocarbons and hydrocarbonaceous materials, such as coal and various petroleum fractions, into hydrogen and carbon black; the steam reforming of coal, petroleum fractions, oil shale, tar sands, lignite, and any other carbonaceous or hydrocarbonaceous feedstock into synthesis gas mixtures, which processes may also include the optional use of one or more inorganic carbonates (such as limestone or dolomite) or inorganic oxides to chemically react with any sulfur-containing contaminants such that they may be removed from the resultant synthesis gas mixtures; the partial dissociation of hydrocarbons and hydrocarbonaceous materials into lower

molecular weight compounds; the partial pyrolysis of saturated hydrocarbons into unsaturated hydrocarbons, such as ethylene, propylene and acetylene; the conversion of organic waste materials, such as sewage sludge or lignin-containing by-products, into a fuel gas, the complete or partial desulfurization of sulfur-containing hydrocarbonaceous feedstocks; the reduction of mineral ores or inorganic compounds to the element or to lower-oxidation-state compounds with a reducing agent, for example, hydrogen, carbon, carbon monoxide or synthesis gas (i.e. a gas mixture comprising carbon monoxide and hydrogen); and the partial or complete reaction of an inorganic element or compound with a carbonaceous material to form the corresponding inorganic carbide.

If desired, one or more catalysts may be used in such high temperature chemical reaction processes to accelerate the reaction or to change its course to a desired reaction sequence. Where such processes involve carbonaceous or hydrocarbonaceous reactants, the addition of an appropriate catalyst to the system may be used to promote the formation of free radicals, carbonium ions or carbanions to influence the course of the reaction.

Of course, no one set of operating conditions is optimum or appropriate for all reactions which may be carried out in the fluid-wall reactor. Operating conditions, such as temperatures, pressures, rates of feed, residence time in the reactor tube, and rates of cooling, may be varied to match the requirements for the particular reaction conducted. By way of illustration, among the factors which influence the products of the pyrolysis of a hydrocarbon are the temperature to which the hydrocarbon is heated and the length of time it is maintained at that temperature. It is known, for example, that methane must be heated to about 2250°F. in order to produce acetylene. Ethylene formation from ethane begins at a lower temperature, about 1525°F. In a typical process for pyrolyzing hydrocarbons, acetylene, ethylene, hydrogen, carbon black, and hydrocarbon oils are produced. Reaction times on the order of a millisecond generally maximize the yield of acetylene, since reaction times of greater than a millisecond generally favor the production of ethylene and other products at the expense of acetylene, while reaction times of less than a millisecond generally reduce the yields of both ethylene and acetylene. Very high temperatures, for example in excess of 3000°F., generally favor the production of carbon black and hydrogen at the expense of acetylene and ethylene. Reaction times in the fluid-wall reactors of the invention may be shortened by shortening the reactor tube and by increasing the rate of flow of reactants introduced into the reactor tube. For very short reaction times, it may be advantageous to mix a radiation-absorbing target, such as carbon black, with the reactants in order to promote efficient coupling between the reactant stream and the thermal radiation from the tube wall and thereby facilitate heating the reactants quickly.

WHAT WE CLAIM IS:—

1. A process for carrying out a chemical reaction at an elevated temperature, wherein one or more reactants are situated in a reaction zone defined by a fluid envelope of generally annular cross-section within a reactor tube, and radiant energy generated outside the reactor tube is collected and focussed onto the reactant or reactants in the reaction zone, the wall of the reactor tube and the fluid envelope both being substantially transparent to the radiation used, sufficient radiant energy being absorbed in the reaction zone to raise the temperature of the or at least one reactant to sustain the chemical reaction.

2. A process as claimed in claim 1, wherein radiation is absorbed by the or at least one reactant.

3. A process as claimed in claim 1 or claim 2, wherein radiation is absorbed by a target within the reaction zone and energy is transferred from the target to the or at least one reactant.

4. A process as claimed in any one of claims 1 to 3, wherein the fluid forming the wall of the reaction zone is inert with respect to the reaction being carried out.

5. A process as claimed in any one of claims 1 to 4, wherein the or each reactant is passed continuously through the reaction zone from an inlet end of the reactor tube to an outlet end thereof.

6. A process as claimed in any one of claims 1 to 5, wherein the annular envelope of fluid is generated by introducing the fluid into the reactor tube through a diffuser situated adjacent to the inlet end of the tube and directing it in a substantially axial direction in substantially laminar flow through the reactor tube.

7. A process as claimed in claim 6, wherein the reactor tube is cooled by the circulation of a fluid coolant substantially transparent to the radiation used.

8. A process as claimed in claim 7, wherein the reactor tube is double-walled and the coolant circulates between its inner and outer walls.

9. A process as claimed in claim 7 or claim 8, wherein the coolant is a gas.

10. A process as claimed in claim 7 or

claim 8, wherein the coolant is liquid or gaseous and is selected from water, heavy water, nitrogen, air and oxygen.

11. A process as claimed in any one of claims 1 to 5, wherein the reactor tube is of porous material and the annular envelope of fluid is generated by causing the fluid to flow radially inwardly through the tube wall.

12. A process as claimed in any one of claims 1 to 11, wherein the radiation is collected and focussed onto the reactant or reactants in the reaction zone by means of a reflector located outside the reactor tube.

13. A process as claimed in any one of claims 1 to 12, wherein the radiant energy is generated by means of a plasma arc, a heated filament or a seeded flame.

14. A process as claimed in any one of claims 1 to 13, wherein the reaction product(s) and any remaining reactant(s) are cooled immediately after they have left the reaction zone.

15. A process as claimed in claim 14, wherein cooling is effected by radiation heat transfer to a cool radiant-energy-absorbing surface.

16. A process as claimed in any one of claims 1 to 15, wherein the or each reactant is additionally heated, at least initially, by means situated within the reaction zone, in order to raise its temperature sufficiently to initiate the reaction.

17. A process as claimed in claim 16, wherein the or each reactant is additionally heated by means of an electrical heating element.

18. A process as claimed in claim 17, wherein the or each reactant is additionally heated by means of an electric arc.

19. A process as claimed in claim 18, wherein the or each reactant is additionally heated by means of a flame.

20. A process as claimed in claim 3 or any claim appendant thereto, wherein the target comprises a target material which is passed through the reaction zone.

21. A process as claimed in claim 20, wherein the target material is introduced into the reaction zone prior to the introduction of the reactants.

22. A process as claimed in claim 20 or 21, wherein the target material is cooled immediately after leaving the reaction chamber.

23. A process as claimed in any one of claims 20 to 22, wherein the introduction of the target is discontinued after the reaction has been initiated.

24. A process as claimed in any one of claims 20 to 23, wherein the target material is a liquid.

25. A process as claimed in any one of claims 20 to 23, wherein the target material is a gas that absorbs electromagnetic radiation having a wavelength within the range of from 0.01 to 100 μm.

26. A process as claimed in any one of claims 20 to 23, wherein the target material is finely divided carbon powder.

27. A process as claimed in claim 3 or any claim appendant thereto, wherein the reaction zone contains a solid target member.

28. A process as claimed in claim 27, wherein the target member is made of carbon.

29. A process as claimed in any one of claims 1 to 28, wherein the power flux in the reaction zone is at least 180 watts/cm^2.

30. A process as claimed in any one of claims 1 to 29, wherein the temperature within the reaction zone is at least 2300°F.

31. A process as claimed in any one of claims 1 to 30, wherein at least one hydrocarbon and/or at least one hydrocarbonaceous material is dissociated into hydrogen and carbon black.

32. A process as claimed in any one of claims 1 to 30, wherein at least one carbonaceous or hydrocarbonaceous material is steam-reformed to yield a gaseous product comprising carbon monoxide and hydrogen.

33. A process as claimed in claim 32, wherein the carbonaceous or hydrocarbonaceous material is selected from coal, petroleum fractions, oil shale, tar sands and lignite.

34. A process as claimed in claim 32 or claim 33, wherein the starting material additionally comprises one or more inorganic carbonates and/or inorganic oxides in order to facilitate the removal of sulphur-containing contaminants from the said gaseous product.

35. A process as claimed in any one of claims 1 to 30, wherein at least one hydrocarbon and/or at least one hydrocarbonaceous material is partially dissociated into lower-molecular weight compounds.

36. A process as claimed in any one of claims 1 to 30, wherein at least one saturated hydrocarbon is subjected to partial pyrolysis to yield one or more unsaturated hydrocarbons.

37. A process as claimed in any one of claims 1 to 30, wherein organic waste material is converted into a fuel gas.

38. A process as claimed in any one of claims 1 to 30, wherein a catalyst for promoting the formation of free radicals, carbonium ions or carbanions is added to reactant or reactants.

39. A process as claimed in any one of claims 1 to 30, in which a sulphur-containing carbonaceous or hydrocarbonaceous material is subjected to partial or complete desulphurisation.

TOTAL P.03

40. A process as claimed in any one of claims 1 to 30, wherein a mineral ore and/or an inorganic compound is reduced by means of a reducing agent to the element or to a compound in which the element is in a lower oxidation state.

41. A process as claimed in claim 40, wherein the reducing agent is hydrogen, carbon, carbon monoxide, or a gas mixture comprising carbon monoxide and hydrogen.

42. A process as claimed in any one of claims 1 to 30, wherein an inorganic element or compound is partially or completely reacted with a carbonaceous material to yield an inorganic carbide.

43. A process for carrying out a chemical reaction at an elevated temperature carried out substantially as hereinbefore described with reference to the accompanying drawing.

44. A product of a high-temperature chemical reaction whenever produced by a process as claimed in any one of claims 1 to 43.

45. A reactor for carrying out a chemical reaction at an elevated temperature, comprising a reactor tube having an inlet end and an outlet end, means for generating an envelope of fluid to define a fluid-walled reaction zone within the reactor tube, a source, located outside the reactor tube, of radiant energy to which the wall of the reactor tube is substantially transparent, and means for collecting and focussing onto the reaction zone, in operation, sufficient radiant energy from the source to raise the temperature of one or more reactants situated there to sustain a chemical reaction.

46. A reactor as claimed in claim 45, which includes means for cooling the reactor tube.

47. A reactor as claimed in claim 46, wherein the reactor tube has an inner wall and an outer wall which together define an annular channel for coolant, the channel having an inlet and an outlet.

48. A reactor as claimed in claim 45 or claim 46, wherein the means for generating an envelope of fluid within the reactor tube comprises a diffuser situated adjacent to the inlet end of the tube and arranged so to admit fluid under pressure to the interior of the tube that the fluid will flow in a generally axial direction through the tube in substantially laminar flow.

49. A reactor as claimed in claim 45, wherein the reactor tube is of porous material and the means for generating an envelope of fluid within the reactor tube comprises means for introducing a fluid under pressure through the porous tube wall to constitute a protective blanket for the inner wall of the tube.

50. A reactor as claimed in any one of claims 45 to 49, which further includes a reflector located outside the reactor tube for collecting radiation from the source and focussing it onto the reaction zone.

51. A reactor as claimed in any one of claims 45 to 50, where the radiant energy source comprises a plasma arc generator, a heatable filament or a seeded flame burner.

52. A reactor as claimed in any one of claims 45 to 51, which includes means situated outside the reactor tube adjacent to its outlet for cooling a product and/or a reactant leaving the reactor tube.

53. A reactor as claimed in any one of claims 45 to 52, which includes additional heating means within the reactor tube for raising the temperature of the reactant or reactants to initiate the reaction.

54. A reactor as claimed in claim 53, wherein the additional heating means comprises an electrical heating element, an electric arc generator, or a flame burner.

55. A reactor as claimed in any one of claims 45 to 54, wherein a radiant-energy-absorbing target member is positioned within the reaction zone.

56. A reactor as claimed in claim 55, wherein the target member is made of carbon.

57. A reactor as claimed in any one of claims 45 to 56, wherein the reactor tube is made of glass, quartz, hot-sintered aluminium oxide or hot-sintered yttrium oxide.

58. A reactor as claimed in any one of claims 45 to 56, wherein the reactor tube is made of an organic polymeric material.

59. A reactor as claimed in any one of claims 45 to 56, wherein the reactor tube is made of an inorganic salt.

60. A reactor for carrying out a chemical reaction at an elevated temperature, substantially as hereinbefore described with reference to, and as shown in, the accompanying drawing.

61. A high-temperature chemical reaction process whenever carried out using apparatus as claimed in any one of claims 47 to 62.

62. A product of a high-temperature chemical reaction process as claimed in claim 61.

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1530315 COMPLETE SPECIFICATION

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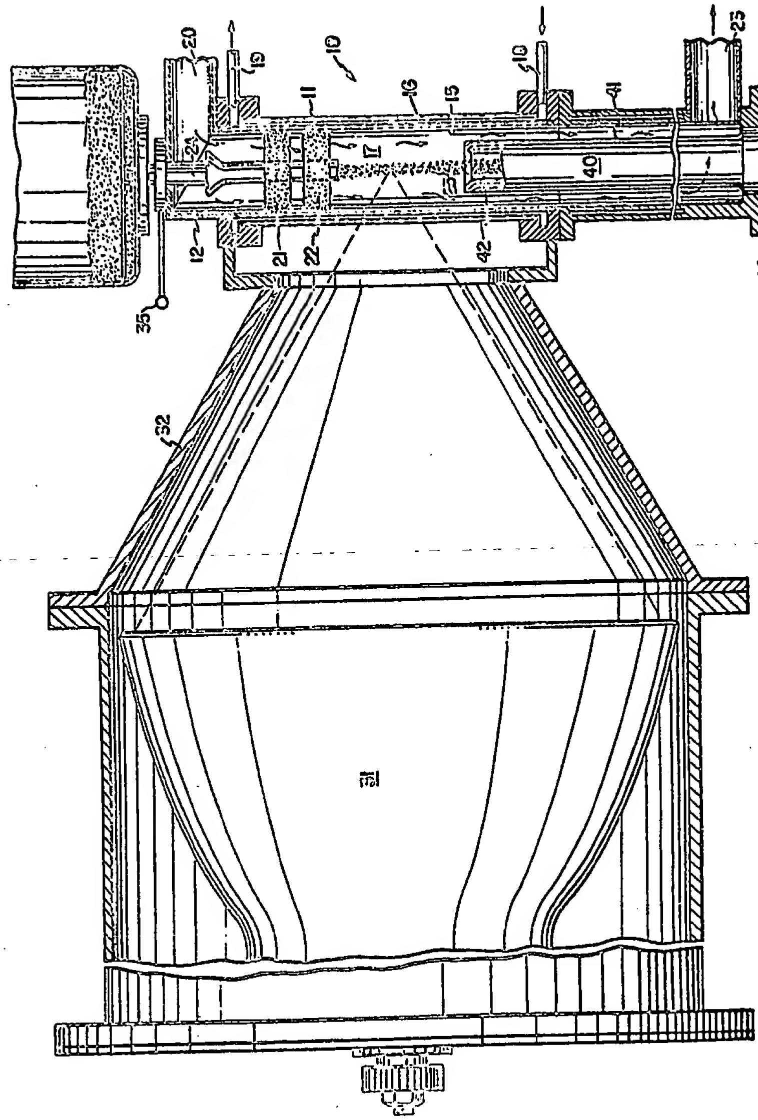

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